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A Concentration Rebound Method For Measuring Particle Penetration And Deposition In The Indoor Environment

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ABSTRACT

Continuous, size resolved particle measurements were performed in two houses in order to determine size-dependent particle penetration into and deposition in the indoor environment. The experiments consisted of three parts: 1) measurement of the particle loss rate following artificial elevation of indoor particle concentrations, 2) rapid reduction in particle concentration through induced ventilation by pressurization of the houses with HEPA-filtered air, and 3) measurement of the particle concentration rebound after house pressurization stopped. During the particle concentration decay period, when indoor concentrations are very high, losses due to deposition are large compared to gains due to particle infiltration. During the concentration rebound period, the opposite is true. The large variation in indoor concentration allows the effects of penetration and deposition losses to be separated by the transient, two-parameter model we employed to analyze the data. For the two houses studied, we found that as particles increased in diameter from 0.1 to 10 μm , penetration factors ranged from ~ 1 to 0.3 and deposition loss rates ranged from 0.1 and 5 h^{-1} . The decline in penetration factor with increasing particle size was less pronounced in the house with the larger normalized leakage area.

INTRODUCTION

Particulate air pollution is associated with increased morbidity and mortality even at the generally low levels of air pollution found in United States cities (Dockery et al., 1993, Pope et al. 1995, Samet et al., 2000). The exact compounds and/or particle size ranges responsible for these health effects have not yet been determined. The indoor environment provides a

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significant, if not dominant, exposure potential for particles for two primary reasons. First, people spend most of their time indoors – typically ~90 % (Jenkins et al., 1992, Robinson and Nelson, 1995). Second, indoor concentrations of particles of outdoor origin are estimated to be on the same order as outdoor concentrations (Wallace, 1996; Ott et al., 2000; Riley et al., 2001). Indoor concentrations of particles of outdoor origin are influenced by many building and environmental factors, such as air leakage rates and ventilation system design. Thatcher et al. (2001) identified and evaluated sources of data for those factors that affect the transport into and concentration of outdoor particles within the indoor environment. In addition to particles of outdoor origin, particles generated from indoor sources, such as tobacco smoke, cooking fumes, or pet dander, may present significant specific health concerns and may add to the total health burden associated with particle exposures.

Particle deposition within the home will reduce indoor airborne concentrations of particles with both indoor and outdoor origins. For this reason, understanding deposition loss rates under typical residential conditions is important for assessing human health impacts from indoor particles. Many experiments have been performed to study particle deposition in the indoor environment (Offermann et al, 1985; Xu et al., 1994; Byrne et al., 1995; Thatcher and Layton, 1995; Fogh et al., 1997; Abt et al., 2000; Long, et al., 2000; Vette et al., 2001; Mosley et al., 2001; and Thatcher et al., 2002). Results from these studies show a wide degree of variability in deposition rate for any given particle size. This variability is due, at least in part, to variations in the conditions under which deposition rates were measured. Factors such as airflow conditions, quantity and nature of furnishings, interior surface-to-volume ratio, surface-to-air temperature differences, particle stability, and measurement method may all be expected to influence the measured deposition rate.

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Another key factor influencing indoor exposures to particles of outdoor origin is the effect of losses due to particle filtration by the building shell. These losses are typically quantified by the use of a penetration factor, P , defined as the fraction of particles in the infiltrating air that pass through the building shell. Previous experiments on penetration factors have found a large variation in values. In some cases, it appears these variations are due to the variability found in houses and in experimental conditions. In other cases, the results are confounded by the inability to separate deposition and penetration effectively and to account for time dependencies in the underlying measured values. Thatcher and Layton (1995) measured particles as a function of size and found penetration factors near 1 for particles with diameters larger than $1\text{ }\mu\text{m}$ for the single residence studied. These results suggested that the shell of the building studied provides essentially no filtration for these particles. However, this study was performed using only a small number of replicates in a single residence and may not be representative of the general building population. Wallace (1996) also calculated penetration factors very close to 1 for PM_{2.5} and PM₁₀, based on the particle mass data from the EPA PTEAM study for a large number of households in the Los Angeles area. These data do not lend themselves to understanding whether particle penetration is particle-size dependent. Cristy and Chester (1981) generated large quantities of $2\text{-}\mu\text{m}$ -diameter spores outside a trailer home, which are often poorly sealed structures, and measured the indoor concentration response. Based on their data, they conclude that penetration losses did not have a significant effect on indoor concentrations. Vette et al. (2001) reported penetration factors for a single house between 0.4 and 0.9 for ambient particles with diameters between 0.01 and $2.5\text{ }\mu\text{m}$. However, they did not measure air exchange rates during the period for which the penetration rates were calculated. Because air exchange rates vary from period to period, their results have a large uncertainty associated with them. Thatcher

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and Layton (1995), Wallace (1996), and Vette et al. (2001) all based their results on measured ambient particle concentrations. Ambient outdoor particles may undergo unknown reactions and/or transformations in the indoor environment that can complicate interpretation of results. As we discuss further below, this is particularly important in the western U.S. where volatile ammonium nitrate particles can represent a significant portion of the particle mass.

Abt et al. (2000) calculated a factor they termed the ‘effective penetration efficiency’, which combines both deposition and penetration losses for outdoor particles. However, their analysis did not separate the two factors. Long, et al. (2001) described an ‘infiltration factor’ which was equivalent to the ‘effective penetration efficiency’ discussed above, where the effects of deposition and penetration losses are not separated. They also determined values for deposition and penetration losses using a random effects mixed model on data from nine homes and reported penetration factors between about 0.9 and 0.3 for particles between 0.02 and 6 μm , respectively. They reported penetration values for one individual home where the “windows and doors were predominantly left open”. As the particles increase in diameter from 1 to 6 μm they report that modeled “penetration efficiencies drop precipitously” ($P = 0.9, 0.82, 0.74, 0.69$, and 0.53 for particle diameter bins 1-2, 2-3, 3-4, 4-5 and 5-6 μm , respectively). The low values for the larger particle sizes seem physically unreasonable, since when windows are open nearly all of the air exchange will occur through the open windows, where the penetration factor will be 1. A potential reason for the incongruity using their approach is that the method fits a single value of penetration factor to all of the data, regardless of whether windows and doors are open or closed. Since homes will tend to have higher air exchange rates (and penetration factors near 1) when windows are open and lower air exchange rates when windows are closed, a single value for P cannot be used to fit the entire data set. Additionally, Long et al. used a steady state model

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to analyze their data. However there is no indication that the indoor concentration was constant over time ($dC_i/dt = 0$), as required by a steady state model or an analysis of the effect of using a steady state model to analyze non-steady data.

Roed and Cannell (1987) reported $P = 1$ for two radioactive isotopes (^{131}I and ^7Be) and $P = 0.53$ for a third (^{137}Cs), all assumed to be bound to particles, based on measurements in a single house. Koutrakis et al. (1992) measured PM 2.5 in 394 homes and estimated penetration factors between 0.58 and 1.04 for 8 elements primarily of outdoor origin. Their calculations assumed that all 8 elements had an average deposition velocity of 0.18 m/h – in effect, that the elements were all associated with the same size particle. If the elements were truly associated with the same particle size, then the reason for the differences in penetration factors between elements is unclear. If they are associated with different particle sizes, then the assumption of a common deposition rate, independent of particle size, will lead to large errors in the calculation of penetration factors. This will be especially true for particles larger than 1 – 2 μm in diameter (Thatcher et al. 2002).

Chao and Tung (2001) report $P = 0.85$ based on measurements of PM 2.5 in five homes and the assumption that indoor deposition losses (β) were negligible. Consequently, their reported penetration factor attributes both penetration and deposition losses to building shell filtration and therefore overstates the penetration loss rate. Tung et al. (1999) measured concentrations of PM 10 in an interior conference room and corridor of an office building under conditions with the HVAC off and calculated penetration factors for transport from the corridor into the adjacent room ranging from 0.69 to 0.86. Their data show that a large increase in the particle concentration in the corridor did not cause any response in the conference room concentration, which they attribute to the low air exchange rate (0.29 h^{-1}) between the corridor and room.

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However, if a transient model for air flow and particle transport between the corridor and the room is constructed using their parameters, the modeled room concentration shows a substantial response to the increase in the corridor particle concentration. A possible explanation for this discrepancy is that some other pathway, such as the HVAC ductwork, duct leaks, or the ceiling plenum, was the main source of particle-bearing air infiltrating into the conference room, not the leaks between the room and the corridor. Furthermore, with the HVAC system off, it is not clear whether the room has a negative pressure with respect to the corridor, which would have to be the case in order to have air flow and particle transport between these two spaces.

McMurry et al. (1985) measured indoor/outdoor (I/O) ratios in a well-sealed residence, with no known indoor sources and found no correspondence between these ratios and particle diameter for particles between 0.1 and 1 μm . In developing their approach and analysis, they assumed that the indoor and outdoor concentrations were at steady state, even though the low air exchange rates and correspondingly long residence times in this study make steady state conditions more difficult to achieve. The potential impact of assuming steady state when the system is transient can be seen in their data during a rainfall episode where the I/O ratios rise dramatically for all size ranges due to a sharp drop in outdoor concentrations. These I/O ratios decline rapidly once the rainfall episode has concluded and outdoor particle concentrations rebound. The changes in the I/O ratio over this period are probably influenced more by the time lag between the indoor and outdoor concentrations than by changes in the physical processes influencing indoor concentrations.

In addition to the whole house studies listed above, several studies have investigated penetration through manufactured cracks in experimental chambers. Lewis (1995) reported penetration factors ranging from 0.97 to 0.29 for particles increasing in size from 1 to 6 μm

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passing through a Perspex (plastic) slit 0.1 mm high and 40 mm wide with a pressure differential of 10 Pa maintained across the slit. Mosely et al. (2001) passed monodispersed particles through manufactured aluminum slits 0.508 mm high and 10 cm wide. They found penetration factors between 0.02 and 0.9 for 2 μm particles and 0.001 and 0.05 for 5 μm particles, with a strong dependence on the pressure differential across the crack.

In general, investigators conducting chamber studies have reported lower penetration rates than those performing whole house studies. The reasons for this are not entirely clear. However, it is reasonable that the penetration factor should be highly influenced by the size and geometry of the infiltration route, which has not been systematically investigated for a wide variety of houses. Liu and Nazaroff (2002) performed experiments measuring penetration factors as functions of particle size, crack height, pressure drop across the crack, and crack material. They found that the height of the crack significantly influenced the penetration factor. For example, they found that for 2 μm particles traveling through a 9.4 cm long smooth crack, essentially no particles deposited in a crack 1 mm high and nearly all particles deposited in a crack 0.25 mm high. Older homes, and other homes which are not tightly sealed, may have a significant portion of their infiltrating air entering through openings around pipes and electrical outlets, poorly sealed windows, and other pathways with relatively large dimensions. The ASHRAE Fundamentals Handbook (1997) reports that a large fraction of air infiltrating into a residence can come from large openings such as those around fireplace dampers (0 to 30%) or in the heating system (3 to 28%). When large pathways are predominant, penetration factors would be expected to be close to unity. In a home without larger openings, where most of the air enters through smaller cracks, the observed penetration rates may approach those found in chamber studies.

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Separating the effects of deposition and penetration in a full-scale house is difficult. Often assumptions are made with respect to either the deposition rate or penetration factor and the resulting parameter is then used to calculate the other factor. Alternatively, both Thatcher and Layton (1995) and Vette et al. (2001) first determined the deposition rate by elevating the indoor particle concentration, measuring the particle loss rate as a function of particle size, and subtracting the exfiltration rate. The measured deposition rates were then used along with ambient indoor and outdoor measurements (with resuspension minimized) to determine the size dependent penetration rate. Using this method, any changes in the deposition loss rate or variability in outdoor particle concentrations at ‘steady state’ will lead to inaccuracies in the calculated penetration factor.

In the present study, we describe a methodology for specifically determining particle penetration as a function of particle size that takes into account the significant factors influencing such measurements, such as the particle size dependent deposition rate onto indoor surfaces, time variation in air exchange rates and outdoor particle concentrations, and the potential effects of other particle loss mechanisms indoors. In contrast to many of the previous studies, we seek to understand the physics of penetration loss mechanisms using actual houses. Since both the chemical composition and physical characteristics of particles vary with size, understanding particle penetration as a function of particle size will be a critical component in testing and evaluating various health and exposure hypotheses. It is also important to understand whether particle penetration factors can be related to the leakage characteristics of houses.

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EXPERIMENTAL PROTOCOL

Since both deposition and penetration losses are particle size dependent and occur simultaneously, it is difficult to decouple the effects of these processes in a residence. In our experimental design the indoor concentration is varied over a wide range and we analyze events where deposition is the dominant loss mechanism and events where penetration losses dominate. A transient model employing a two-parameter fit is used to determine the combination of deposition rate and penetration factor that best fits the observed data.

For this study we conducted experiments in two houses, one in Clovis, and the other in Richmond, California. At both sites, a typical experiment began with a short period of indoor resuspension activities followed by a 1 to 2 hour period of concentration decay. For some experiments at the Clovis house, a natural-gas-stove burner was also ignited to increase the indoor concentration of smaller particles. Following the concentration decay period, the buildings were then pressurized using a High Efficiency Particle Absolute (HEPA) filter mounted in a window at the locations shown in Figures 1 and 2. The HEPA filter supplies essentially particle free air (greater than 99.97% removal of 0.3 μm diameter particles) and creates a positive pressure gradient across the building shell (indoor pressure greater than outdoor) that prevents the entry of outdoor particles, causing the indoor particle concentration to decline to nearly zero. The effectiveness of the pressurization was confirmed by measuring the indoor-outdoor pressure differential across the building surfaces at various points. When the HEPA filter was then turned off, infiltration of particle laden outdoor air resumed, and the indoor particle concentration rebounded. During the entire experiment, tracer gas measurements were made continuously at several locations within the building. Size-differentiated particle concentrations were measured every 3 minutes with particle instruments located both inside and

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outside the building.

The resuspension process raises indoor concentrations of particles larger than $\sim 0.2 \mu\text{m}$ in diameter by causing particles that have been deposited or tracked onto surfaces to disperse in the indoor air. It is expected that these resuspended particles will be chemically stable in indoor air and will not undergo transformation processes. During the concentration decay period, indoor concentrations are relatively high and losses due to deposition will be large compared to gains due to particle infiltration. The deposition rates obtained during this period are stable and reproducible. During the concentration rebound period, the opposite will be true. Changes in indoor particle concentrations will be due almost exclusively to infiltrating outdoor particles and deposition losses will be small compared to infiltration. Thus we can effectively separate penetration and deposition losses. In some cases, chemical transformation can be an important confounder; we discuss this further below.

Study Locations

To explore the effect of building characteristics on deposition losses and penetration factors, we applied this method in two separate homes, each with very different construction. Experiments were performed in two houses: one in Richmond, California and a second in Clovis, California. The Richmond house is a small, older, single-story building (59 m^2) with wood clapboard siding, un-insulated walls, and double-hung wooden windows. The building is located in a relatively unsheltered location near the San Francisco Bay and is subject to winds that exhibit a significant diurnal variation in both wind speed and direction, as well as occasional high wind speeds. Figure 1 shows a floor plan of the house. All particle measurement equipment was located in the living room and two oscillating fans sitting on the floor were used

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to promote mixing, as shown in Figure 1. The building contained a wall heater that was not operated during these experiments.

The Clovis residence is a moderate sized home (134 m^2) constructed in 1972. It has a stucco exterior and single-glazed aluminum frame windows. The house is single story, with standard height ceilings (2.4 m), a forced air heating and cooling system (which was not operated during these experiments), and ceiling fans (which were operated during the experiments to promote mixing). An additional oscillating fan located approximately 1.5 m from the floor was operated in the living room to disperse tracer gas and promote mixing near the particle measurement equipment. The house is located in a residential suburb of Fresno, California, surrounded by mature trees and homes of a similar height and size. The flat terrain and high level of sheltering resulted in relatively low levels of wind loading near the building. Figure 2 shows a floor plan of this house, along with the location of equipment.

Blower door measurements, in which the amount of airflow under various imposed pressure differentials is measured, are commonly used to compare the relative 'air tightness' of homes (ASHRAE Standard 136, 1993). These measurements can be used to calculate normalized leakage (NL), which is the leakage area normalized by floor area and a house height factor. The average house in the United States has a NL of $1.2 \text{ cm}^2/\text{m}^2$ (Sherman and Matson 1997). The typical NL of a conventional new house is approximately $0.55 \text{ cm}^2/\text{m}^2$ and a new well-sealed, energy-efficient house is around $0.31 \text{ cm}^2/\text{m}^2$ (Sherman and Matson, 2002). Older homes tend to have significantly larger NL than newer homes (Sherman and Dickerhoff, 1998). The Richmond house has a normalized leakage area around $2.5 \text{ cm}^2/\text{m}^2$, indicating that the structure is fairly 'leaky' and that air penetrates the building easily. The Clovis house has a relatively low normalized leakage area of $0.65 \text{ cm}^2/\text{m}^2$, indicating that it is better sealed and may have a smaller

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proportion of the air leakage occurring through large penetrations in the building shell.

Measurement Equipment

The indoor particle and gas measurement instruments were located in the living room of each house. Systems to measure tracer gas concentration and pressure differentials across the building shell monitored the living room as well as several locations throughout the house, as shown in Figures 1 and 2. An additional set of particle measurement devices was also located outdoors at each house. In Richmond, the outdoor instruments were located on a table approximately 30 feet upwind of the house. In Clovis, the instruments were located in a shed adjacent to the house and samples were collected through a PM 10 inlet projecting approximately 10 feet above the shed roof.

Two aerodynamic particle counters (TSI, Model APS 3320), one located indoors and one outdoors, were used to measure size distributions for particles with aerodynamic diameters between 0.5 and 10 μm . To assure that the measurements from the two instruments were comparable, measurements with the instruments sitting side-by-side were performed before and after each test series. The comparison tests were performed both outdoors and indoors. The results were compared to determine a collection efficiency ratio between the instruments for each size bin. For a typical calibration series, the standard deviation of the collection efficiency ratio was less than 0.1 for the individual particle size bins. The efficiency ratios were used to adjust the concentrations obtained by the instruments and ‘match’ the results. For the Clovis experiments, the matching experiments were performed with the outdoor instrument sampling through the PM 10 inlet manifold so that manifold losses would be included in the correction factor. The APS uses 52 size bins between 0.5 and 20 μm . For our analysis, the smallest size bin

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and all size bins over 10 μm were eliminated, because of poor correlation between instruments for the smallest size bin and the use of the PM 10 inlet outdoors reduced or eliminated particles with diameters larger than 10 μm . Data in the remaining bins were grouped to produce 12 size bins between 0.5 and 10 μm . The concentration in the largest of these bins was often very low indoors particularly in the Clovis house with its low air exchange rates. For each size bin, we excluded periods where there were insufficient counts in the bin to provide adequate count statistics from our analysis, typically around 5 counts per measurement interval.

Two optical particle counters (Particle Measurement System, Model LAS-X), one located indoors and one outdoors, were used to measure size distributions for particles with optical diameters between 0.1 and 3 μm . The instruments were ‘matched’ in a manner analogous to that used for the aerodynamic particle counters with similar precision in the collection efficiency ratio. The smallest and largest size bins were excluded due to difficulty in matching and a low number of counts indoors for the larger sizes. Calibrations using an Electrostatic Classifier (TSI Inc, Model 3071) were performed with ambient particles to determine the appropriate conversion from optical to aerodynamic size. However, the conversion from optical to aerodynamic diameter is highly dependant on particle composition and varies over time. In particular, variations in the chemical composition of the aerosol, such as high levels of carbonaceous aerosol, will lead to errors in the conversion from optical to aerodynamic diameter. As a result, we report our concentrations in terms of optical diameter for measurements performed with the optical counters. The optical diameter is not necessarily expected to be identical to the aerodynamic diameter for the particles studied.

In Clovis, CA, a substantial and highly variable fraction of the ambient aerosol can be composed of ammonium nitrate particles. These particles have been shown to dissociate into

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ammonia and nitric acid in the indoor environment (Lunden et al. 2002) and therefore cannot be readily modeled with a simple physical model that assumes that the particles are conserved. To determine the contribution of ammonium nitrate particles to the outdoor aerosol during these experiments, PM_{2.5} nitrate, carbon and sulfate were measured with 10-minute resolution using the integrated collection and vaporization method of Stolzenburg and Hering (2001). This method collects PM_{2.5} particulate matter by humidification and impaction onto a 1 mm diameter spot on a metal substrate. The sample is then analyzed by flash-vaporization and quantitation of the evolved vapor compounds. Nitrate concentrations are measured using low-temperature vaporization in a nitrogen carrier gas with quantitation of the evolved vapors using a chemiluminescent monitor equipped with a molybdenum converter to reduce higher oxides of nitrogen to nitric oxide. Sulfate and carbon analyses are performed using high-temperature heating, with analysis of the evolved sulfur dioxide by UV-fluorescence and carbon dioxide by nondispersive infrared absorption.

Indoor and outdoor measurements were performed simultaneously using a four-cell system. One pair of cells (one indoors and one outdoors) was used for nitrate measurements. A second pair was used for the combined measurement of carbon and sulfate. The outdoor nitrate cell and outdoor sulfate-carbon cell were housed indoors inside a box that was ventilated with outdoor air to maintain near-outdoor temperature at the point of sample collection. A more detailed description of this system can be found in Lunden et al. (2002).

Air infiltration rates were measured using sulfur hexafluoride tracer gas techniques (ASHRAE, 1997) with a photo-acoustic infrared detector system (Brüel and Kjær, Model 1312). For the Richmond experiments, an injection/decay method was used. In this method, a pulse of tracer gas is injected and mixed within the space. The loss of tracer gas over time is then

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monitored at three to six locations and the concentration decay rate is used to determine the infiltration rate. During the Clovis experiments, a constant injection system was used. In this method, tracer gas is injected at a constant rate before and during each experiment. The time varying infiltration rate is calculated using a transient mass balance approach that accounts for the injection rate and the time dependent infiltration losses. In general, the calculated air exchange rate at a given time was essentially the same for each measurement location within the house and the average concentration for all locations was used in the model. The only exception occurred when the window-mounted HEPA filter was operated in the Clovis residence. During these periods, the SF₆ concentration in the room containing the HEPA fan was very low and that measurement point was excluded from the average in order to obtain an air exchange rate more representative of the entire house.

An automated, multiple channel differential pressure system (Energy Conservatory, APT 8) was used to monitor pressure differentials across the building shell at various points for each house (see figures 1 and 2) and to assure that pressurization with the HEPA filtration system was always positive (inside with respect to outside) on all surfaces of the building envelope.

Analysis Method

The concentration of particles indoors is a balance between the sources and sinks of particles in the indoor environment. Figure 3 illustrates potentially important factors affecting indoor concentrations. In the most general form, the indoor concentration of particles of a specific size and composition particle can be represented by the following equation.

$$\frac{\partial C_I}{\partial t} = (C_O P - C_I) \lambda_v - C_I \beta + G + S + F + K + H \quad (1)$$

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Where: C_I = indoor particle concentration at time t ($\# \text{ cm}^{-3}$),
 t = time (h),
 C_o = outdoor particle concentration at time t ($\# \text{ cm}^{-3}$),
 P = penetration factor,
 λ_v = air exchange rate (h^{-1}),
 β = deposition loss rate (h^{-1}),
 G = generation of particles indoors ($\# \text{ cm}^{-3} \text{ h}^{-1}$),
 S = particle formation through gas-particle conversion ($\# \text{ cm}^{-3} \text{ h}^{-1}$),
 F = particle formation due to chemical reaction ($\# \text{ cm}^{-3} \text{ h}^{-1}$),
 K = particle size change through coagulation ($\# \text{ cm}^{-3} \text{ h}^{-1}$), and
 H = particle size change through hygroscopic growth ($\# \text{ cm}^{-3} \text{ h}^{-1}$).

For the conditions and particle size ranges used in these experiments, we do not expect coagulation (K), hygroscopic growth (H), or formation (F) to have a significant impact on indoor particle concentrations. In addition, care was taken to avoid indoor sources (G) by using an unoccupied space and operating equipment that could generate particles, such as pumps, outdoors. Therefore, we assume that these effects are negligible in our analysis. During some experiments, observations indicated that the results were complicated by dissociation and vaporization of ammonium nitrate particles (S). Periods of high ammonium nitrate were excluded from our results. For the periods selected for our analysis, we could reduce the mass balance equation to:

$$\frac{\partial C_I}{\partial t} = C_o P \lambda_v - C_I (\lambda_v + \beta) \quad (2)$$

Under steady-state conditions, this equation is solved easily for a constant outdoor concentration and stable infiltration rate. However, in most real-world situations – as was the case for these experiments – there is significant time variation in C_o and λ_v causing the steady-state solution to be invalid. When $\partial C_I / \partial t$ is not zero, the equation can be solved using a basic ‘forward-marching’ scheme with time step, Δt , as shown below:

$$C_I(t_2) = C_I(t_1) + PC_o(t_1)\lambda_v(t_1)\Delta t - C_I(t_1)(\lambda_v(t_1) + \beta)\Delta t \quad (3)$$

In these experiments, the time step used was 3 minutes, corresponding to the measurement interval for the size resolved instruments.

The equation solving methodology employed the minimization of the sum of the square of the differences between the measured and modeled indoor concentration at each time step divided by the measured concentration at that time. Since the concentration varied over several orders-of-magnitude during each experiment (from a maximum after resuspension to a minimum during the filtration period), dividing by the measured concentration normalized the weighting of each point over the entire experiment. Although this leads to biases due to measurement errors, it was found to provide better fits over the entire data range than produced by other weighting schemes. For instance, without weighting the high concentrations periods dominated and the curve fit at lower concentrations was poor. A weighting scheme minimizing the inverse of the measured/modeled difference led to large errors at high concentrations. Individual measurements which appeared to be inaccurate due to either equipment limitations or

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instabilities were excluded from the analysis if they exhibited the following characteristics: (1) zero or near zero counts or (2) one or more points where counts differ by more than 50% from the counts in both the preceding and following measurements in the time series, with no physical explanation. In general, the excluded measurements represented less than 1% of the total number of points, with the exception of the largest size channel of each instrument, which exhibited very low particle counts over significant periods.

The Excel spreadsheet tool SOLVE (Microsoft Corporation) was used to determine the deposition loss rate, β , and penetration factor, P , which minimized the sum of the weighting function residuals for each particle size range. Due to bias introduced by weighting the model/measurement error by the measured concentration, the average modeled concentration tended to be slightly lower than the measured average concentration. This difference was typically less than 5 %. In consideration of this bias, the goodness-of-fit was determined using two measures: the first is the measured/modeled correlation coefficient and the second is the difference between the measured and modeled average concentration. If the correlation coefficient was less than 0.95 or the difference between the average concentrations was more than 10% for a time series reconstruction for a given particle size range, then the fit was rejected and the results were discarded. Typically the model performed very well and few rejections were observed, for example, in the Richmond experiments, 12 of the 122 reconstructions were rejected, and only 3 of the 81 reconstructions were rejected in the Clovis data. One experiment performed at the Clovis house was discarded due to problems associated with interference by ammonium nitrate, which will be discussed in more detail subsequently. For each particle size range, the deposition loss rates and penetration factors obtained from the reconstructions were averaged over all experiments performed at each home. The average and standard deviation are

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reported for each site.

RESULTS AND DISCUSSION

The penetration rebound method produces a characteristic indoor particle profile as shown in Figure 4 for smaller particle sizes measured with the optical particle counter and Figure 5 for larger particles measured with the aerodynamic particle counter. Immediately following resuspension, indoor concentrations are significantly higher than during the quiescent periods at the end of each experiment, for all but the smallest particle sizes. Concentrations of particles greater than $0.5\ \mu\text{m}$ typically increase by more than a factor of 5 and particles larger than $3\ \mu\text{m}$ increased by more than a factor of 10 compared with the quiescent background. Similarly, during the period when the house was pressurized with filtered air, indoor concentrations are significantly lower than quiescent concentrations. This large range of indoor concentrations increases the sensitivity of the method and improves the ability to find unique solutions for the penetration factor and deposition rate. However, the use of resuspended and natural-gas stove generated particles as sources of additional particle concentrations indoors may introduce error into the calculations if the properties of these particles (such as shape and density) are different than those found in ambient air. To the extent that resuspended particles represent materials originally deposited from air, they should be similar to ambient particles but lacking the most volatile or unstable components. The portion of resuspended particles originating from tracked materials may not represent the chemical composition of ambient air well, but as long as they are stable and similarly shaped the deposition behavior should be similar to that observed for ambient particles. Mechanically separated materials, such as fibers, are typically larger than $10\ \mu\text{m}$ and would not be included in the measurements. Particles from natural gas combustion

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should be similar to those in other fresh combustion aerosols, but they may not represent other submicron particles, such as inorganic aerosols, well if the densities are significantly different.

Changes in the outdoor concentration during the experiment increase the sensitivity of the fit because deposition losses are a function of indoor concentration and penetration losses are a function of outdoor concentrations. Consequently, outdoor concentration variability helps differentiate the effects of penetration and deposition losses. Significant changes in outdoor concentration occur over the course of one or more hours on a typical day, but they can occur more rapidly, for instance when the wind direction shifts. For particle sizes near the larger end of an instrument's range, low indoor particle counts often lead to poor count statistics and a high degree of sample-to-sample variability especially during the rebound phase or when air exchange rates are low. This variability can lead to a decrease in the sensitivity of the fit of the model to the data. Figures 4 and 5 illustrate the data and model fit during the experiments. The model fits well on both the downward sloping deposition phase and on the upward sloping rebound phase, indicating that both deposition and penetration losses are appropriately described.

The sensitivity of the model fit to the penetration factor value can be seen in Figure 5. This example uses APS data from the Richmond experiments with the model constrained to a penetration factor arbitrarily fixed at 0.5 leaving the deposition rate as the sole fitting parameter. The fixed penetration factor results in poorer fits and reduced correlation coefficients (from 0.994 to 0.965 for 0.9 - 1.1 μm , from 0.996 to 0.973 for 2.1 – 2.6 μm , and from 0.993 to 0.988 for 5.0 – 6.2 μm). In this example, where the penetration factor is lower than the 'best-fit' penetration factor, the solver reduces the modeled deposition rate to compensate for the artificially low penetration rate. This results in an over-prediction of the indoor concentration in the initial period just following resuspension, when deposition is the dominant loss mechanism.

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Later in the experiment, when indoor concentrations are lower, the effect is reversed, with the model under-predicting indoor concentrations. If the penetration factor were fixed above the 'best-fit' value, the opposite would be observed. These results demonstrate that constraining the penetration rate creates a situation where it is not possible to adequately fit both the concentration decay and rebound portions of an experiment.

At the Clovis site, the effects of ammonium nitrate dissociation and vaporization indoors confounded the effects of penetration factor and deposition rate on indoor concentrations. Figures 6 and 7 illustrate this effect. As shown in Figure 6, the outdoor ammonium nitrate concentrations are low and the modeled particle concentrations match the measured particle concentrations well early in the experiment. Later, when the outdoor nitrate concentrations rise rapidly, the outdoor particle counts show a similar rise for particles in size bins between 0.3 and 1 μm , but exhibit little effect for larger or smaller particles. This suggests that outdoor ammonium nitrate particles have diameters primarily between 0.3 and 1 μm . The model predicts that indoor particle concentrations will increase substantially when the outdoor particle concentrations increase, as expected from equations 2 and 3. However, indoor measurements of indoor particle nitrate concentration and indoor particle concentrations do not show corresponding responses. This indicates that deposition and penetration losses are not the only processes affecting the indoor particle concentrations at this site during times of high outdoor ammonium nitrate concentrations. As a consequence, a simple chemical-conservative model cannot be used to represent adequately the indoor particle concentration during these periods. Methods used for assessing losses of particle nitrate and conversion to gaseous ammonia and nitric acid indoors are described in more detail by Lunden et al (2002) and Fischer et al (2002).

Determining the size distribution for ammonium nitrate and subtracting the nitrate particles

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from the size-resolved particle concentrations both indoors and outdoors could remove the effect of nitrate transformation. Since our time-resolved nitrate measurements are not size-resolved, this method would require a very stable nitrate size distribution. Figure 7 shows indoor and outdoor particle concentrations for selected size ranges and PM_{2.5} nitrate concentrations measured both outdoors and indoors during a period when no rebound experiments were conducted. Initially the outdoor PM_{2.5} nitrate concentration is low and stable. Between approximately 0900 and 1200, a nitrate peak occurs outdoors. The outdoor particle size distributions for particles with diameters less than about a micron exhibit peaks during the nitrate episode. The response appears first in the 0.15 to 0.2 μm size bin and shifts to larger size ranges as the episode develops. By the time the outdoor PM_{2.5} nitrate concentration reaches a maximum, the peak is seen predominantly in the 0.65 to 0.80 μm size bin. Based on the response seen in the outdoor size distribution, the average size of the PM_{2.5} nitrate particles appears to increase as the nitrate peak episode progresses. These sizes are consistent with the bimodal ambient nitrate distribution reported by John et al (1990). The shifting nitrate size distribution makes it difficult to remove the outdoor ammonium nitrate particles from the size distribution, in the absence of size resolved nitrate measurements. The indoor size distributions indicate no corresponding increase in concentration during the nitrate peak episode, suggesting that either the ammonium nitrate aerosols are dissociating very rapidly indoors or they aren't being transported across the building shell. This latter seems unlikely, as other particles in the apparent size range of the ammonium nitrate particles have penetration factors greater than ~ 0.8 .

Due to the difficulties caused by ammonium nitrate dissociation and vaporization indoors, we chose to calculate penetration factors and deposition rates for the Clovis house using data only from those portions of the experiments where the outdoor concentrations of ammonium

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nitrate were low and relatively stable. Despite this constraint, ammonium nitrate may still exhibit a somewhat confounding effect on the results because overall particle concentrations were also low during these periods. This may result in artificially low penetration rate predictions for submicron particles in the Clovis experiments. Since the indoor particles during the deposition portion of the experiments consist mainly of resuspended and/or generated particles, calculated deposition rates are less likely to be affected by ammonium nitrate losses.

Table 1 lists experimental parameters for each of the experiments performed. Figure 8 illustrates how particle concentrations and conditions change over the course of an experiment. The best fit parameters and standard deviations for penetration factor and deposition loss rate at various particle sizes are listed in Table 2. Figures 9 and 10 compare the deposition loss rates and penetration factors obtained at the two sites. The optical diameter values for the Richmond site represent the best fit achieved by applying the two parameter transient model to the data for five experiments conducted during March and April 2000. The aerodynamic data are based on only four of these experiments because the equipment failed during one of the experiments. The values for the Clovis site represent the best fit for three experiments conducted during August, September, and October 2000.

Deposition rates for the Richmond and Clovis houses are similar and fall within the range of deposition rates found by other researchers (Thatcher and Layton, 1995; Fogh et al, 1997; Mosley et al, 2001). The deposition loss rates at the Clovis residence were slightly lower than those observed for the Richmond building for all particle sizes, but the differences were not statistically significant when individual points are compared. Deposition rates can vary between residences and within a single residence for many reasons. Thatcher et al. (2002) showed that both the indoor air speed (related to the internal mechanical energy) and the amount of

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furnishings influence deposition loss rates. In these experiments, the fans used to improve mixing indoors are expected to increase deposition rates over those that would be found in the absence of fans. Consequently, the deposition rates reported here are more representative of homes using fans or forced air heating and cooling than those without. During periods when the window-mounted HEPA filter operated, deposition may have been further enhanced in the room in which the HEPA system was located. However, the air exchange rate is high during HEPA filter operation and the relative contribution of enhanced deposition to the overall particle loss rate is expected to be small. Moreover, the duration of the filter operation when particle concentrations are high (and thus total particle losses are potentially important) is short, approximately 15 to 20 minutes compared with the total time of the experiment, ca 5-6 hrs. Thus the possible effect of enhanced deposition due to the operation of the HEPA filter system is quite small. Both indoor air speed and furnishing level varied between the 2 homes. Mean air speed may also have varied between experiments at the same home due to changes in surface to air temperature differences.

Penetration factors at the Richmond house were near unity for all but the largest and smallest particle sizes. Since it is an older building with a relatively high normalized leakage area, and is constructed with double hung wooden windows and wood siding, the Richmond house is presumed to have larger cracks through the building shell. Large cracks are not expected to be very efficient for removing particles (Liu and Nazaroff, 2001). Penetration rates for the Clovis house were lower than those at the Richmond house, ranging from ~ 0.8 for particles 0.2 to 0.5 μm to just over 0.3 for particles 5 μm and larger. This may mean that in the tighter construction of the Clovis house, the penetration pathways are smaller and more effective for removing particles. However, there are no published studies on the correlation, or lack of

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correlation, between normalized leakage area and average leakage crack dimensions. For particles between 0.1 and 1.0 μm there may be a confounding effect due to ammonium nitrate dissociation in the Clovis house, since ammonium nitrate particles are in this size range—see Figure 7 and John et al (1990), even though the data sets were selected to avoid periods of high nitrate measurements. This may yield calculated penetration factors that are lower than the actual penetration factor for particles in this particular size range.

The correlation between the deposition rates calculated by the two sets of instruments (APS and LAS-X) is very good for most sizes, especially considering that the instruments measure particles in a different manner (aerodynamic and optical diameter, respectively). The standard deviations for the deposition loss rates averaged about 60% of the deposition rate value. It is not clear from this study how much of this variation is due to measurement and fitting inaccuracies and how much is due to actual variability in the deposition rate. A comparison of previous research has indicated that deposition loss rates can vary considerably based on conditions within the interior space (Thatcher, et al. 2002). The standard deviations for the penetration factors averaged 16% of the penetration factor and once again the portion of the variability attributable to experimental uncertainty as opposed to variations in the actual derived rate is unclear. The data and model of Liu, et al. (2001 and 2002) and the experiments of Mosely et al. (2001) suggest that the penetration factor is a function of pressure differential across the crack (reflecting flow rate effects within the building envelope). It is not unreasonable to assume that the penetration factor in real homes will vary with environmental conditions, as these variations induce time variations in the differential pressure across the building shell. The parameter fit is not as sensitive to penetration factor as it is to deposition rate, since the decay period - which provides the best fitting data for deposition losses- is typically longer than the rebound period -

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which provides the best fitting data for the penetration rate, and therefore provides more data points for the model fit. Consequently, more model fit uncertainty is expected for the penetration factor. This is especially true in the upper size channels of both instruments, which sometimes record only a few particles during a measurement period, leading to noisy data and poor count statistics that reduce the accuracy of the reconstructions.

CONCLUSIONS

Deposition and penetration losses are both important factors influencing indoor particle concentrations and resulting exposures. Since these factors operate simultaneously in the indoor environment, it can be difficult to separate their effects experimentally. We have demonstrated an effective method for determining size resolved penetration factors and deposition loss rates in full-scale homes. The two houses studied represent two distinctly different portions of the housing stock and air leakage spectrum; from a leakier, older style building with wood siding construction to a newer style building with tighter construction. Although deposition loss rates were similar for the two houses, penetration factors were significantly different. The older home had high penetration factors (near 1 for most particle sizes) while the newer home showed significant filtration by the building shell (penetration factors near 0.3 for particle larger than 5 μm). Deposition rates were slightly, but not significantly, higher in the older house. Deposition rate differences can be caused by differences in indoor air speed and/or differences in surface to volume ratio.

The composition of the ambient aerosol was shown to have a significant impact on the results and their interpretation as determined from some of the field experiments where ammonia nitrate concentrations were particularly large. Changes in the physical environment and gas

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phase contaminant concentrations indoors can lead to transformation processes, such as ammonium nitrate dissociation, which confounds the interpretation of experiments designed to quantify aerosol losses that result from crossing the building shell. Care must be taken to identify and account for the impact of these processes, in order to avoid improperly attributing their effects to deposition or penetration losses. Consideration and quantification of the various loss mechanisms must also be accounted for when estimating exposure.

The method demonstrated here permits the determination of deposition and penetration with minimal ambiguities. However, we applied the technique to only two houses. Additional work is required to obtain a better understanding of the range, variability, and particle size dependence of the penetration factor in a representative range of real homes. This information will be especially important in evaluating potential causal relationships between specific particle characteristics, such as size, and health outcomes.

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Table 1: Air exchange rates and concentration bounds for selected median particle diameters particle for each experiment.

Location	Date	Time		Air Exchange Rate (/hour)		Maximum Indoor Concentration (particles/liter)			Minimum Indoor Concentration (particles/liter)			Maximum Outdoor Concentration (particles/liter)			Minimum Outdoor Concentration (particles/liter)		
		Start	End	HEPA ^a	Range ^b	0.2µm ^c	1.5µm ^d	4.5µm ^d	0.2µm	1.5µm	4.5µm	0.2µm	1.5µm	4.5µm	0.2µm	1.5µm	4.5µm
Richmond	28mar00 ^e	07:39	11:27	6.8	0.3-0.8	4,300			100			23,600			6,300		
Richmond	28mar00	12:12	15:21	6.3	1.5-3.6	10,800	800	80	260	20	1	15,000	1,300	90	8,000	700	30
Richmond	05apr00	10:24	16:12	5.8	0.9-2.6	84,000	2,000	380	2,000	20	1	173,000	1,600	70	48,000	800	30
Richmond	06apr00	09:54	12:57	5.3	1.9-3.6	142,000	1,400	70	3,200	100	4	89,000	2,000	140	27,000	1,500	60
Richmond	10may00	09:12	11:54	5.7	1.1-1.9	10,600	1,300	380	590	20	1	10,800	750	70	5,000	50	1
Clovis ^f	29aug00	11:36	20:03	2.7/4.6	0.1-0.4	340,000	4,100	350	9,700	20	1	485,000	1,400	90	211,000	210	20
Clovis	28sep00	14:15	18:36	3.5	0.5-0.6	200,000	3,600	90	14,000	50	1	320,000	490	100	260,000	320	50
Clovis	29sep00	10:18	16:48	2.7	0.5-0.6	200,000	6,500	160	14,000	50	1	460,000	750	110	240,000	310	30
Clovis	02oct00	07:39	17:24	2.5	0.3-0.3	92,000	5,700	400	2,500	10	1	320,000	1,600	100	100,000	500	20

- Average air exchange rate when window mounted HEPA filter is operating.
- Range of air exchange rates during experiment, excluding period when HEPA filter is operating.
- 0.2µm particles, as measured using an optical particle counter.
- 1.5µm and 4.5µm particles, as measured using an aerodynamic particle counter
- Only optical particle counter data was available for this experiment.
- During this experiment, there were 2 separate periods of HEPA filtration at 2 different fan speeds.

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Table 2: Average model fit parameters and standard deviations for deposition loss rate and penetration factor at the Richmond and Clovis residences by (a) median optical diameter and (b) median aerodynamic diameter.

a) Optical Particle Counter Results

Median Optical Diameter of Size Range													
Clovis	0.13	0.175	0.225	0.275	0.35	0.575	0.725	0.9	1.125	1.375	1.75	2.25	2.75
Deposition Loss Rate (hr-1)	0.12±0.09	0.11±0.10	0.15±0.16	0.2±0.2	0.2±0.2	0.12±	0.2±0.1	0.2±0.1	0.2±0.1	0.3±0.2	0.4±0.2	0.5±0.2	0.7±0.3
Penetration Factor	0.8±0.2	0.8±0.1	0.6±0.1	0.8±0.3	0.6±0.2	0.8±0.1	0.8±0.1	0.7±0.2	0.8±0.1	0.9±0.1	0.9±0.1	0.9±0.1	0.8±0.1
Richmond	0.13	0.175	0.225	0.275	0.35	0.575	0.725	0.9	1.125	1.375	1.75	2.25	2.75
Deposition Loss Rate (hr-1)	0.3±0.3	0.3±0.3	0.2±0.2	0.2±0.2	0.2±0.2	0.2±0.1	0.4±0.3	0.5±0.4	0.5±0.2	0.9±0.6	0.7±0.2	0.9±0.5	1.1±0.1
Penetration Factor	0.8±0.2	0.93±0.08	1.00±0.08	1.0±0.1	1.0±0.1	1.1±0.2	1.01±0.06	1.0±0.1	1.0±0.1	1.0±0.2	0.9±0.2	0.9±0.2	0.87±0.07

b) Aerodynamic Particle Counter Results

Median Aerodynamic Diameter of Size Range													
Clovis	0.65	0.81	1.0	1.2	1.5	1.9	2.4	2.9	3.6	4.5	5.6	7	8.7
Deposition Loss Rate (hr-1)	0.1±0.1	0.1±0.1	0.1±0.1	0.2±0.1	0.3±0.1	0.5±0.1	0.9±0.2	1.3±0.4	1.8±0.4	2.2±0.8	3.3±0.6	3.5±1.1	3.3±1.3
Penetration Factor	0.7±0.2	0.8±0.2	0.9±0.2	0.9±0.2	0.7±0.04	0.7±0.02	0.62±0.06	0.5±0.1	0.4±0.1	0.4±0.1	0.33±0.05	0.35±0.09	0.48±0.16
Richmond	0.65	0.81	1.0	1.2	1.5	1.9	2.4	2.9	3.6	4.5	5.6	7	8.7
Deposition Loss Rate (hr-1)	0.5±0.7	0.4±0.4	0.4±0.2	0.5±0.4	0.7±0.3	1.0±0.4	1.3±0.5	1.5±0.7	2.3±0.6	2.6±1.0	3.7±0.9	4.8±0.7	5.06 ^a
Penetration Factor	1.0±0.3	1.0±0.2	1.0±0.1	0.93±0.06	0.94±0.05	0.93±0.06	0.93±0.09	0.86±0.03	0.9±0.1	0.9±0.1	0.9±0.1	0.77±0.06	0.88 ^a

a The acceptance criteria for this particle size was met for only one experiment.

FIGURE CAPTIONS

Figure 1: Floor plan of the experimental house located in Richmond, California. Particle measurement equipment was located in room 1. Tracer gas concentrations were measured at locations denoted by stars and indoor-outdoor pressure differentials were measured at the locations shown by the open circles.

Figure 2: Floor plan of the experimental house located in Clovis, CA showing locations of ceiling fans, tracer gas measurements, and differential pressure measurements. Particle measurement equipment was located in the living room and in the outdoor shed.

Figure 3: Schematic of particle transport, transformation and removal processes in the indoor environment.

Figure 4: Measured and modeled indoor particle concentrations for several optical particle diameter ranges during a typical run in Richmond, CA. The larger particle size ranges show increased sample-to-sample variability due to the low number of counts per sample period.

Figure 5: Measured and modeled indoor concentrations for selected aerodynamic particle diameters during a typical experiment at the Richmond site. For the constrained model, the penetration factor is fixed at 0.5 and the deposition rates are determined using the same residual minimization process as used in the unconstrained results.

Figure 6: Measured and modeled indoor concentrations for selected optical particle diameter size ranges, along with outdoor PM_{2.5} nitrate concentrations at the Clovis, CA site.

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The increase in outdoor PM_{2.5} nitrate does not result in corresponding increases in indoor particle concentrations, as would be predicted by the simple physical model that assumes that particles do not undergo phase change.

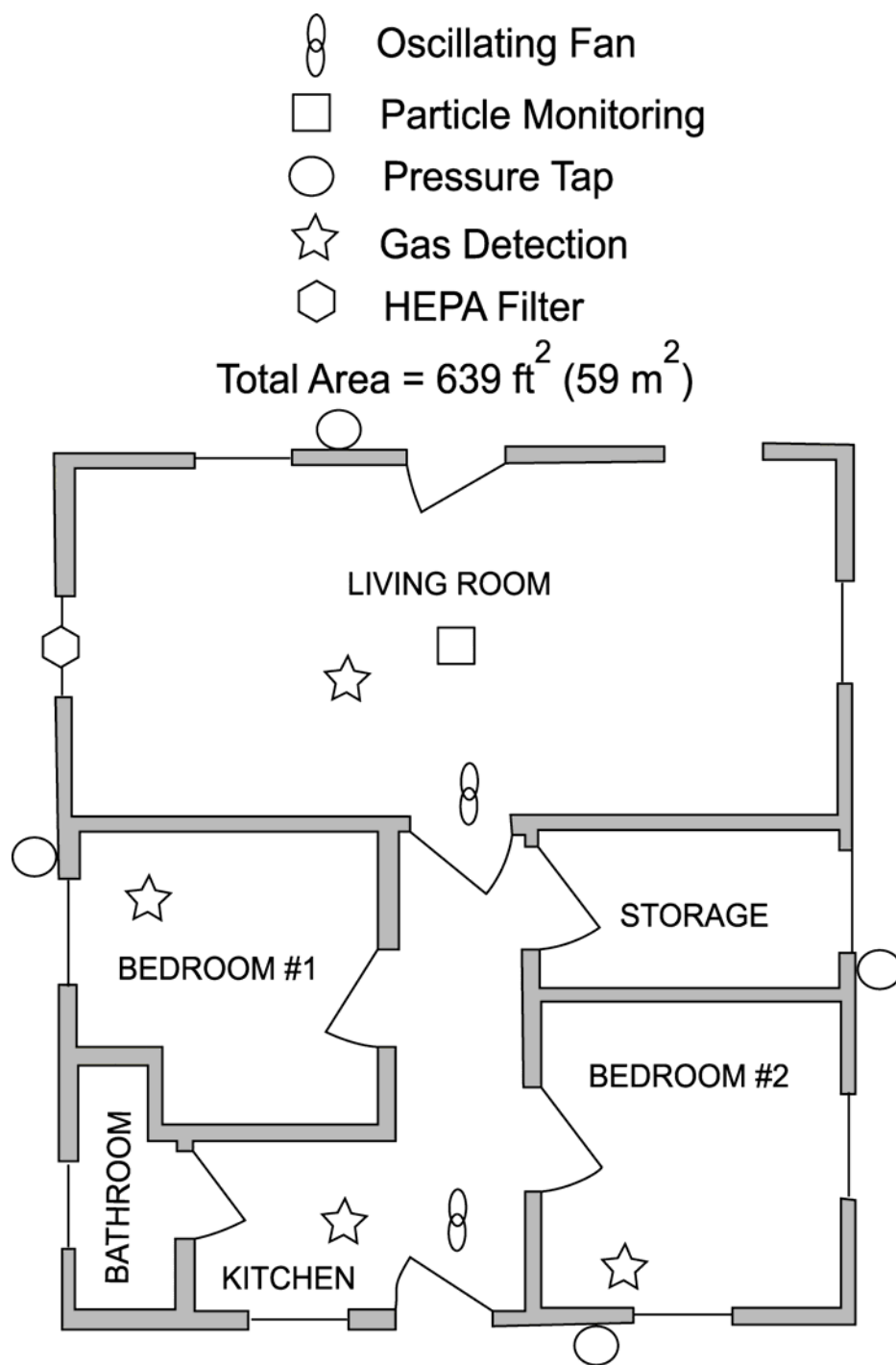
Figure 7: Indoor and outdoor concentrations for PM_{2.5} nitrate and selected optical particle diameter size ranges. The outdoor nitrate peak results an increase in the outdoor particle concentration for particles with optical particle diameters between 0.1 and 1.0 μm , but does not result in a corresponding increase in indoor concentrations.

Figure 8: Air exchange rate and indoor and outdoor temperature, relative humidity, pressure difference and concentrations for selected particle sizes as a function of time over the course of a typical experiment.

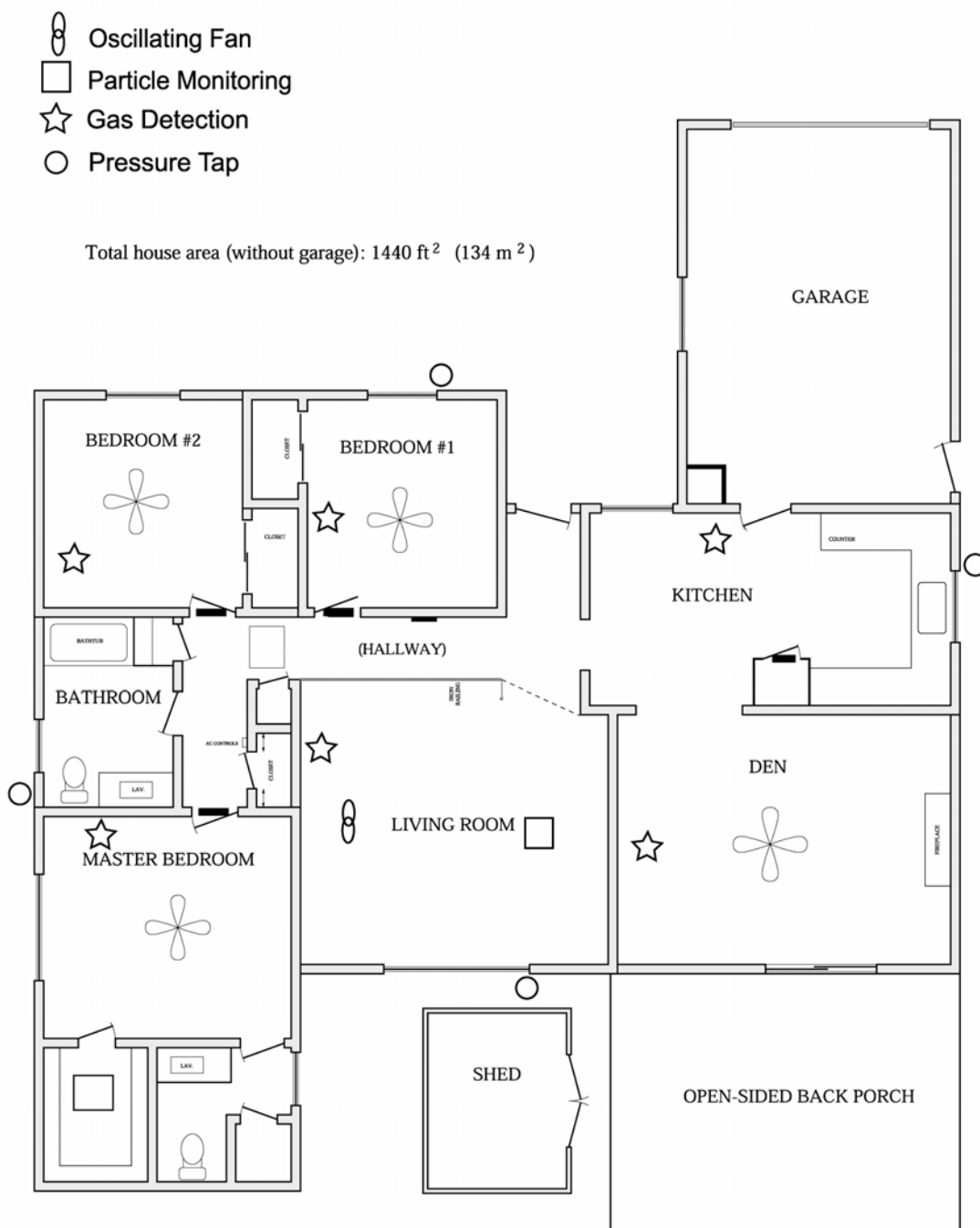
Figure 9: Averages and standard deviations of the parameter fits at the Richmond site for (a) penetration factor and (b) deposition loss rate as a function of optical and aerodynamic particle diameter. The optical and aerodynamic diameters are not necessarily identical for a given particle.

Figure 10: Averages and standard deviations of the parameter fits at the Clovis site for (a) penetration factor and (b) deposition loss rate as a function of optical and aerodynamic particle diameter. The optical and aerodynamic diameters are not necessarily identical for a given particle.

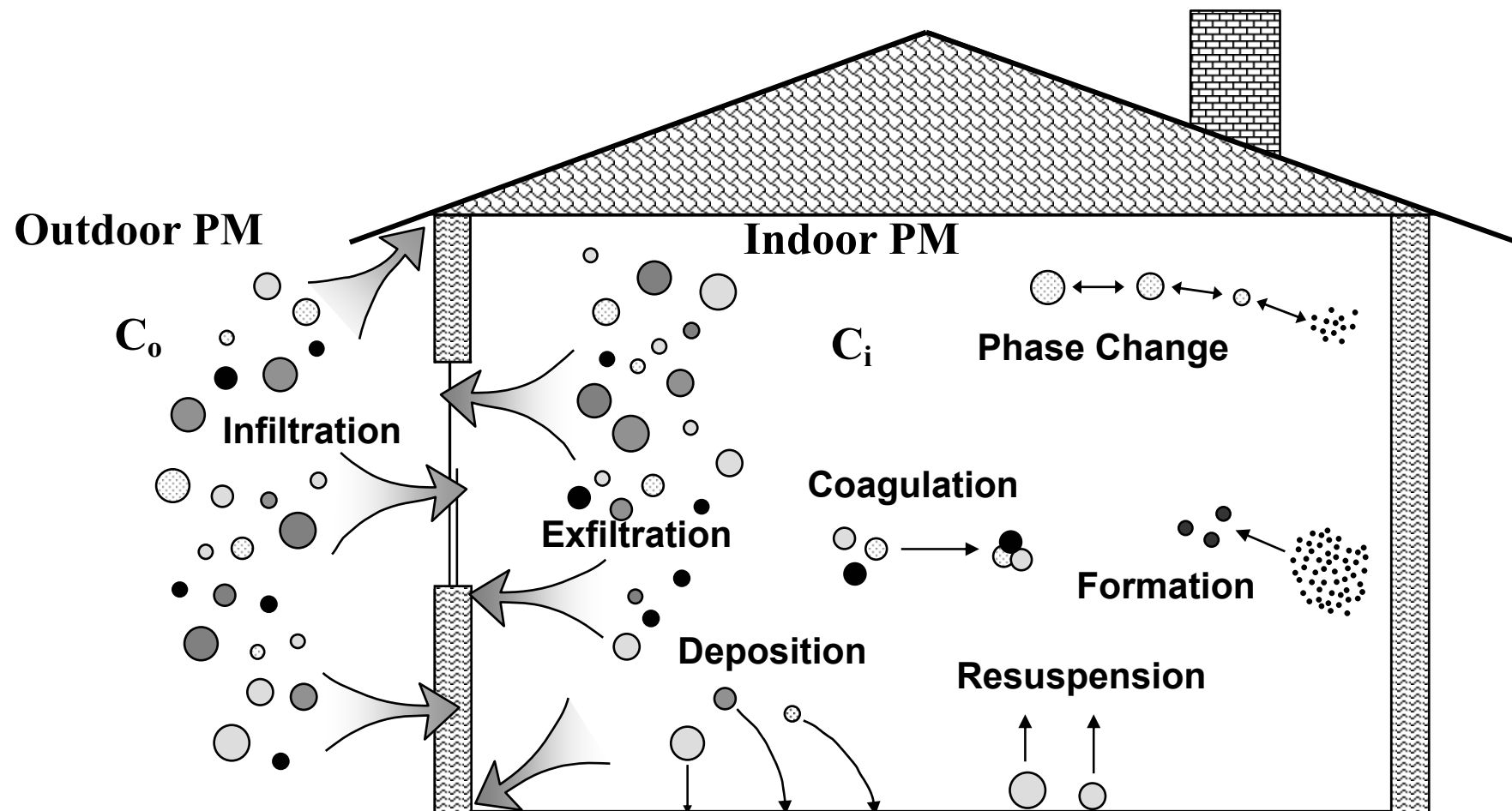
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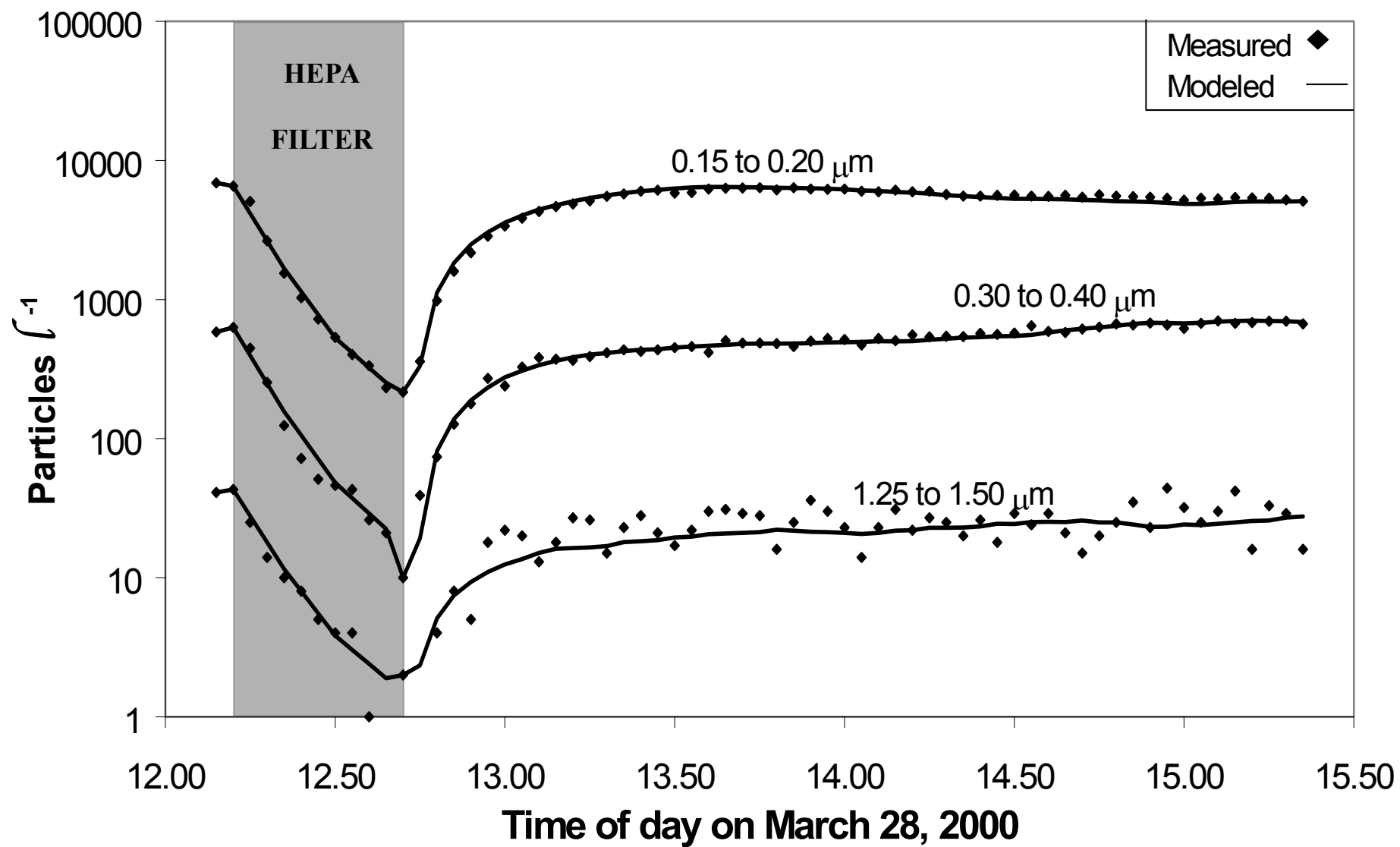


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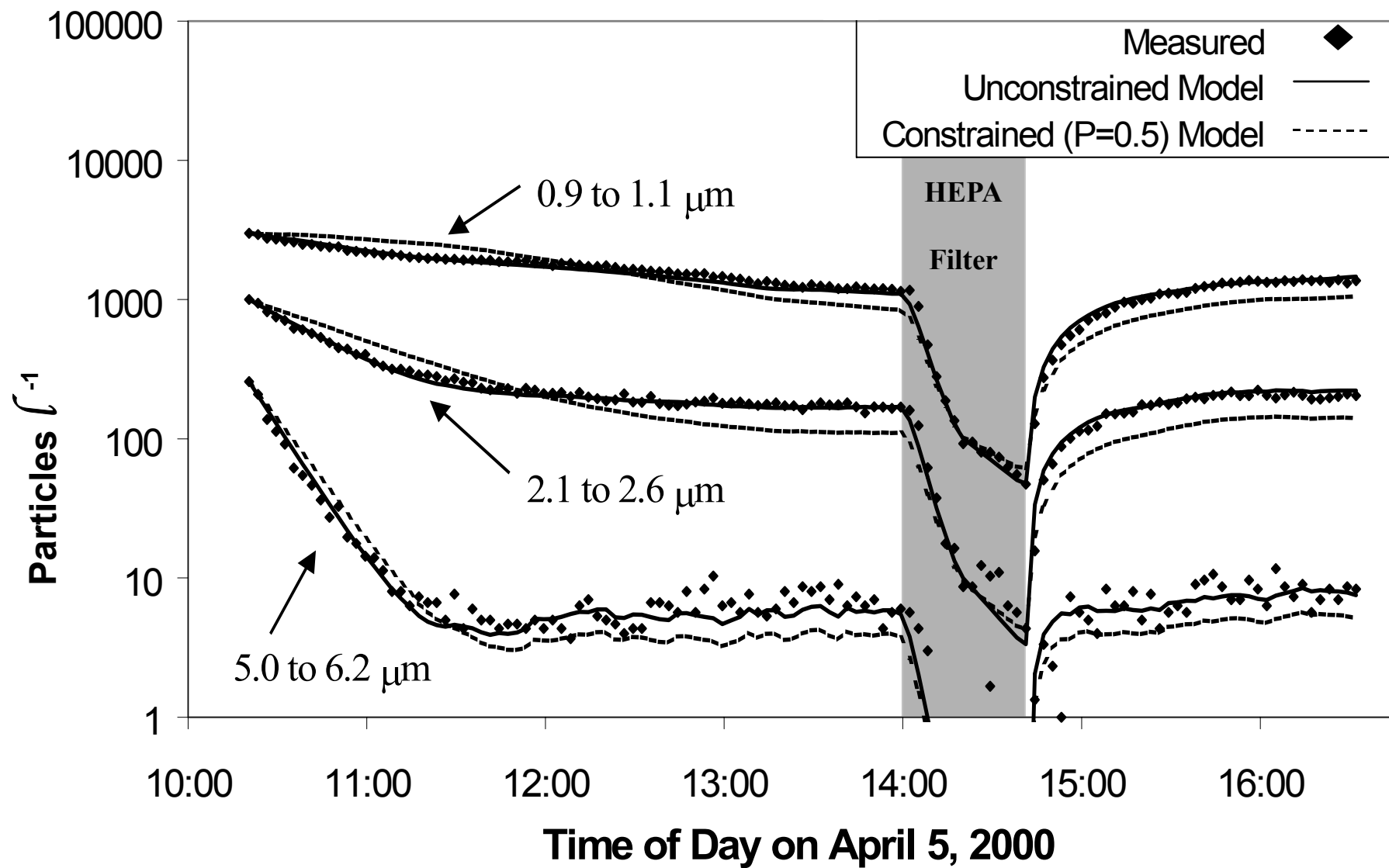


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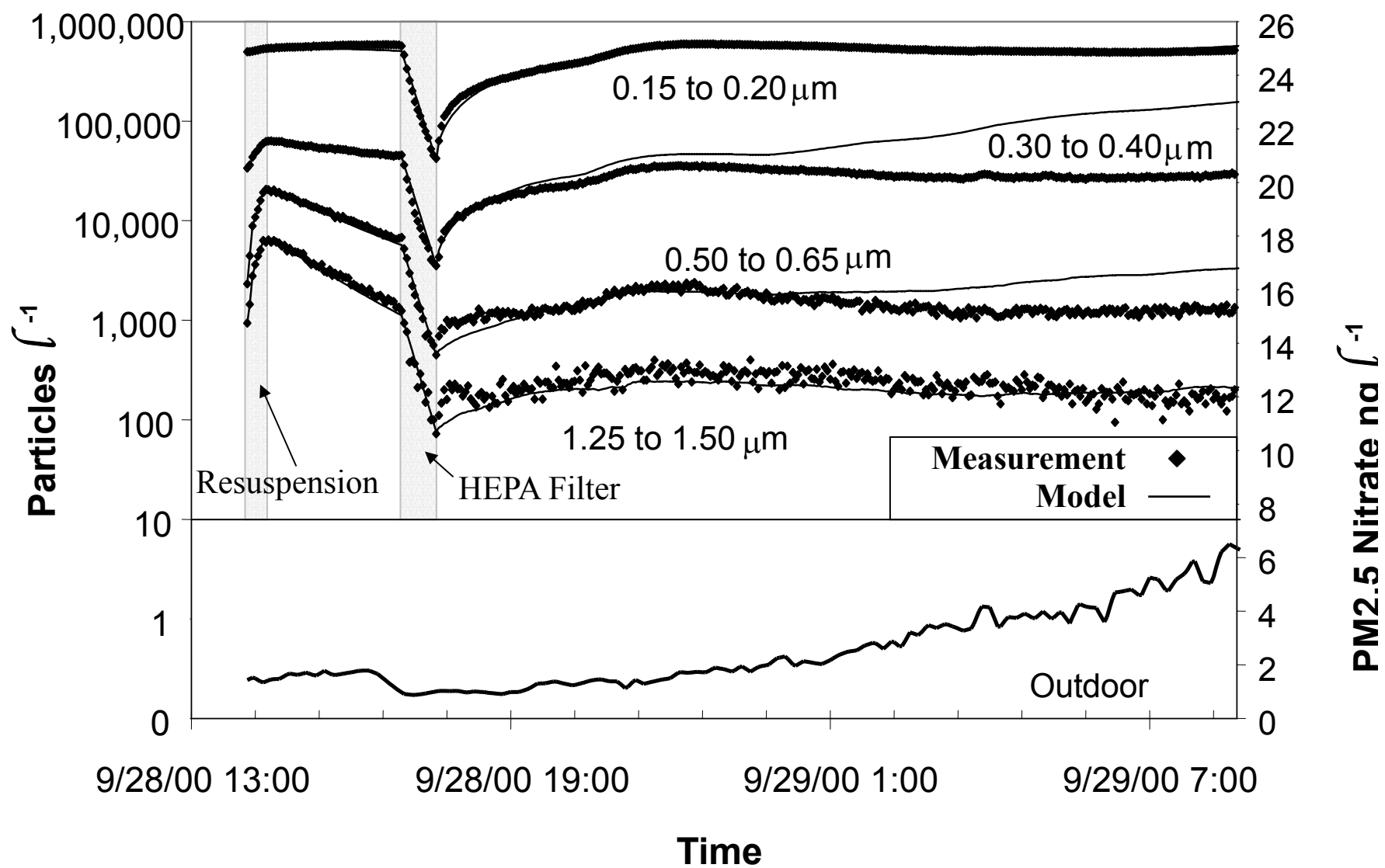




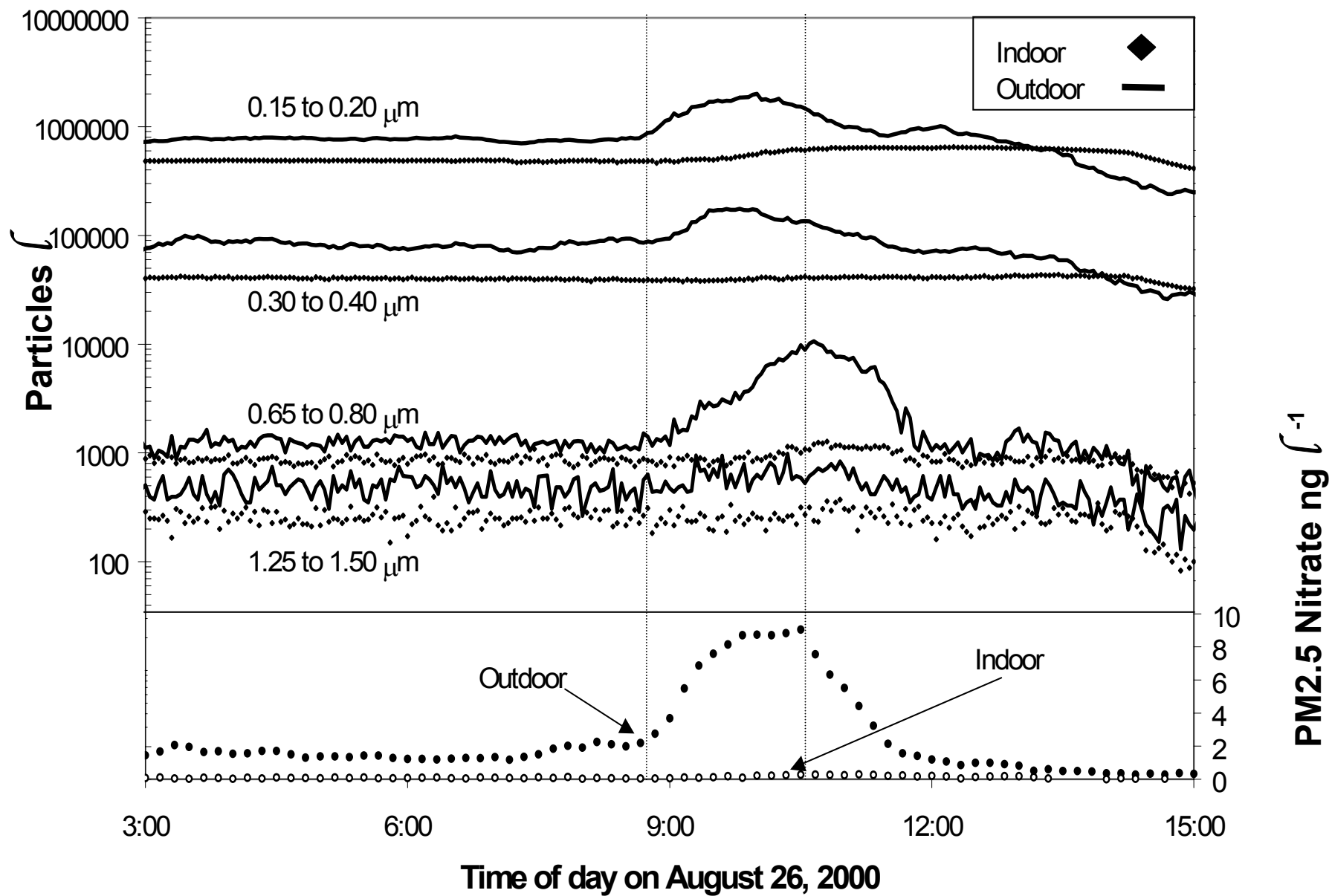
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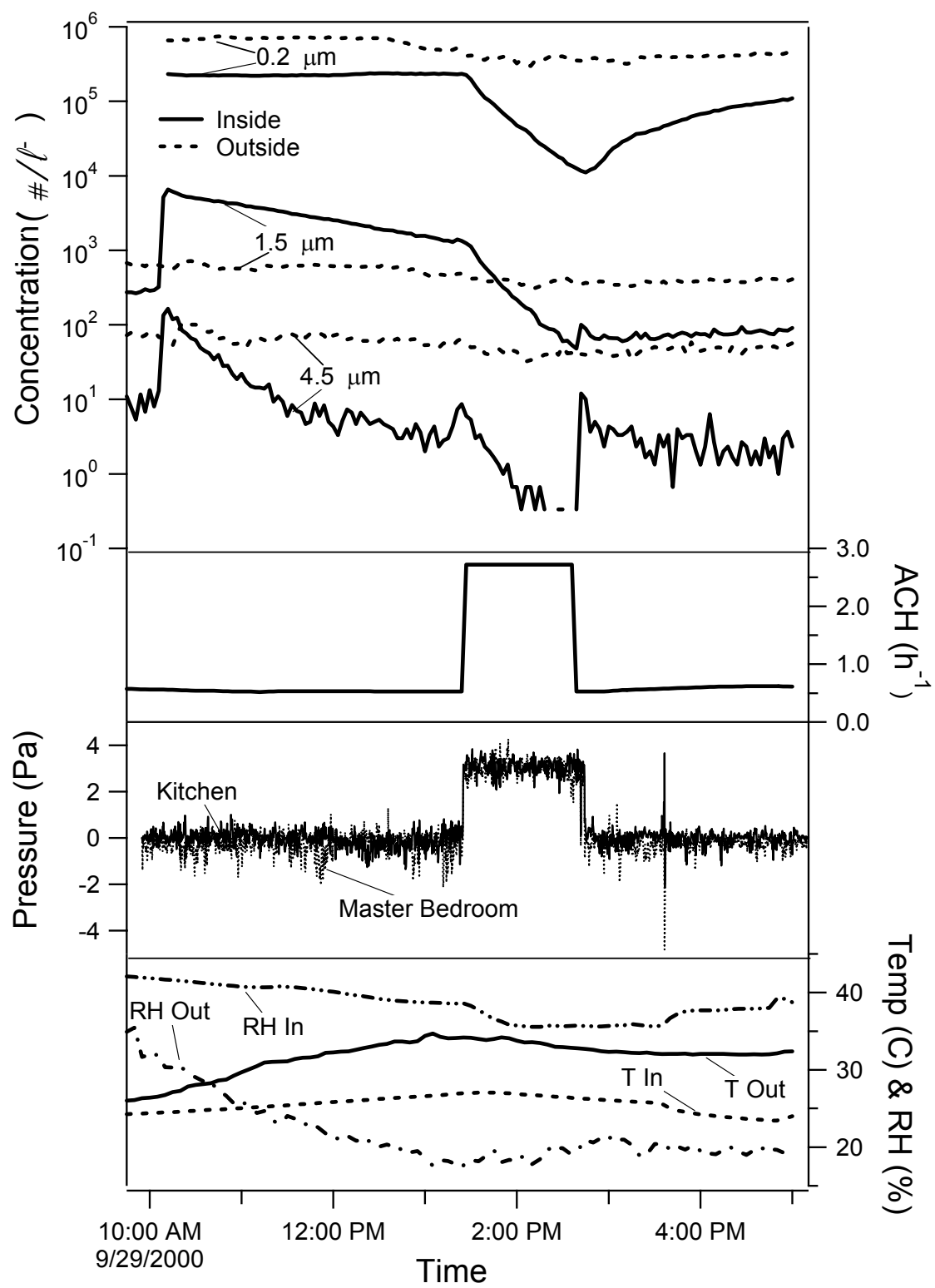
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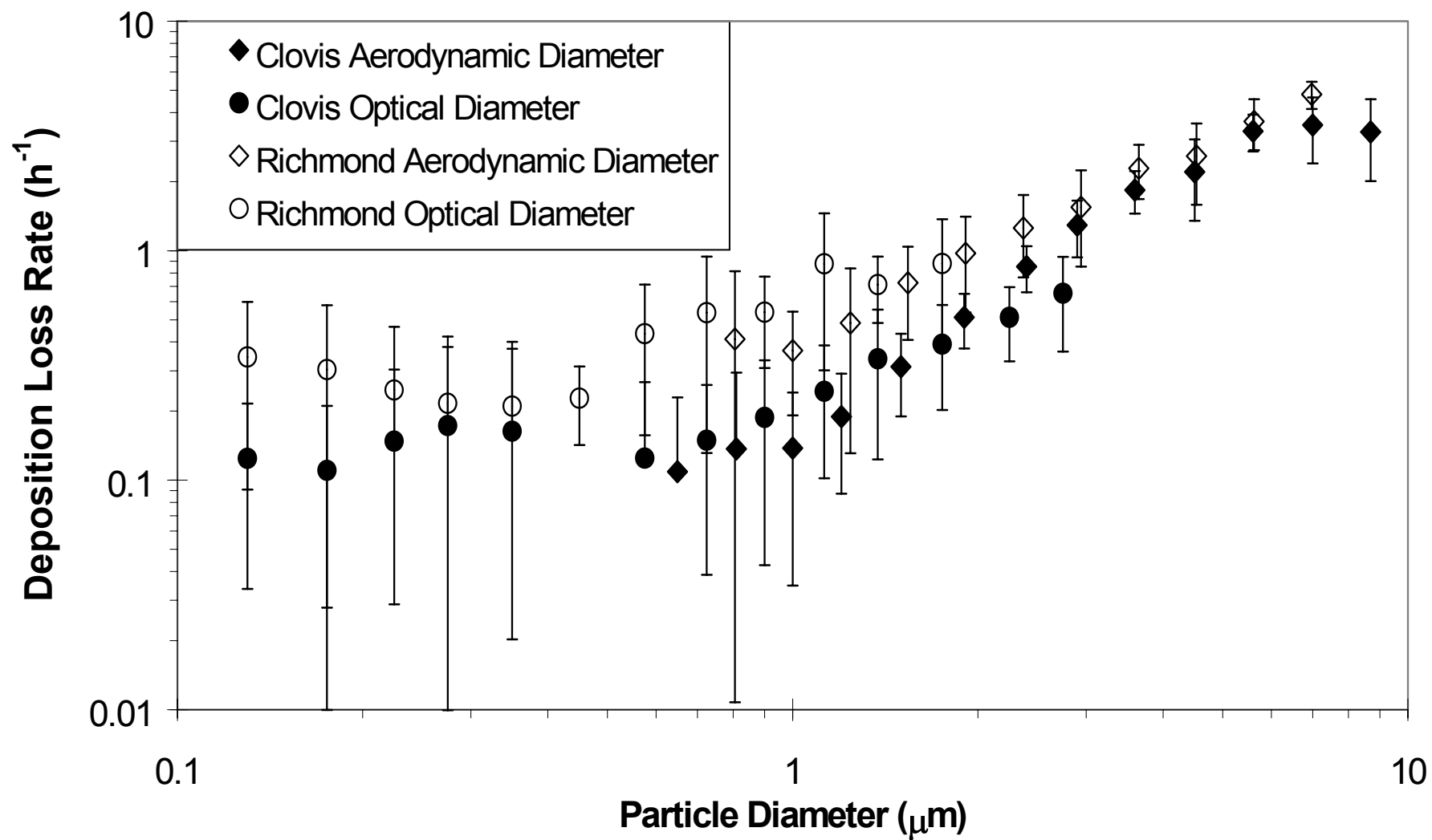
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